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Supported ceric ammonium nitrate: A highly efficient catalytic system for the synthesis of diversified 2, 3-substituted 2,3-dihydroquinazolin-4(1*H*)-ones

Someshwar D. Dindulkar [a,b], Jeongsu Oh [a], Veenita M. Arole [b], Yeon Tae Jeong [a,*]

[a] *Department of Image Science and Engineering, Pukyong National University, Busan 608-737, Republic of Korea*
[b] *Jawaharlal Nehru Engineering College, Aurangabad 431 003, Maharashtra, India*

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ABSTRACT

A practically expeditious protocol has been developed for the cascade synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones via the condensation of 2-aminobenzamide/2-aminobenzanilide and aromatic aldehydes using a catalytic amount of silica-supported ceric ammonium nitrate. This method affords rapid transformation at room temperature with good to excellent yields.

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1. Introduction

Quinazoline heterocycles are very well-known six-membered heterocyclic ring molecules that possess wide biological properties, such as antitumor, antidefibrillatory, analgesic, diuretic, antihistaminic, vasodilating, tranquilizing and antianxiety ones (Fig. 1) [1–9]. Also quinazolines are oxidized into quinazolin-4(3*H*)-ones moieties that are, as growth inhibitors, of great importance in the treatment of leukemia cells, and are also used as poly(ADP-ribose)polymerase-1 inhibitors [10,11]. Also quinazoline derivatives were found to be useful as fungicides, bactericides, insecticides, and plant-growth regulators [12]. Some of them display good pharmacological properties, such as sedative, anticholinesterase, hypotensive, soporific, antispasmodic, tranquilizing, muscle relaxing, antirheumatic, diuretic, antimalarial ones, as well as other activities [13,14].

Several methods have been reported for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one. Among them, the general method includes the reductive cyclization of aldehydes or ketones with 2-aminobenzamide in the presence of acid catalysts, such as cyanuric chloride, $ZrCl_4$, cerium ammonium nitrate, PPA–SiO_2, and gallium triflate [15–19]. Very recently, Ramesh et al. reported efficient methodologies for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones using the three-component aniline/isatoic anhydride/aldehyde system and also the two-component aldehyde/anthranilamide system catalyzed by β-cyclodextrin as a reusable catalyst [20,21]. Also there are some methodologies that were reported recently using two-component or three-component systems in aqueous solutions, ionic liquids, and organic media at high temperature [22–26]. Also Wang et al. reported a manual grinding technique using CAN catalyst, which has some limitations, such as long reaction times, high temperatures, and, practically, the fact that the yield and time needed for manual grinding is variable because of grinding inconsistency [17].

Despite most of the reported protocols give good yields, there are some limitations associated with the reaction

* Corresponding author.
E-mail address: ytjeong@pknu.ac.kr (Y.T. Jeong).

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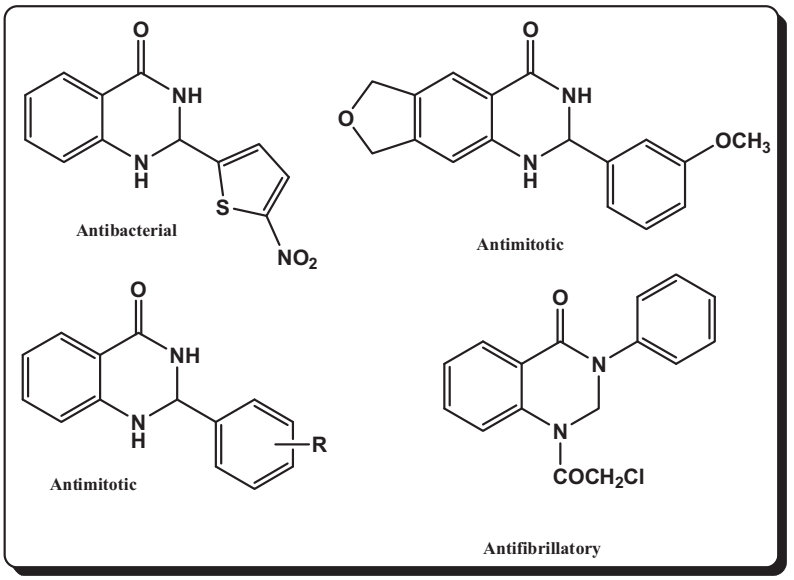


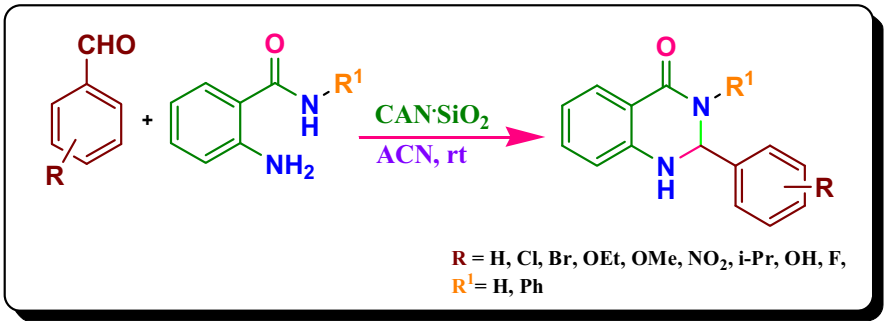
Fig. 1. Reported biologically active quinazolinone and quinazolinone-based molecules.

system, such as prolonged reaction times, high temperatures and the need for high catalyst loading. Therefore, the development of an efficient, cost effective and high-yield protocol for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones is of great interest, with significant demand from the medicinal industries.

We decided to investigate the efficiency of the supported metal Lewis acid catalyst for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones. With this aim, we synthesized a variety of silica-supported heterogeneous catalysts for this one-pot synthesis, and we established an efficient methodology for the cascade synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones from aldehydes and 2-aminobenamide/2-aminobenzanilide using CAN·SiO_2 as an expeditious catalyst at ambient temperature, with good yields (Scheme 1).

Cerium (IV) ammonium nitrate (CAN) is a versatile catalyst, which has been widely used in organic transformations due to its many advantages, such as high reactivity, ease of storage, low cost and commercial availability. The use of this reagent for numerous transformations involving C–C, C–O, C–N, and C–S bond formation has been described [27–29]. However, the application of CAN (ceric ammonium nitrate) is limited due to its poor solubility in common organic solvents. Therefore, adopting silica as the supporting material has been reported as a good alternative [30–33]. Silica gel-supported ceric ammonium nitrate (CAN·SiO_2) proximates the reactant, which fastens electron-transfer processes between reactants, and further enhances the rate of reaction with higher efficiency and thus with shorter reaction times. Moreover, the use of a heterogeneous metal Lewis catalyst instead of traditional homogeneous metal Lewis and Brønsted acid catalysts could be a more environmental friendly alternative.

In connection with our ongoing research on cost effective methodologies, we discovered a new reaction system for the synthesis of a variety of organic compounds of therapeutic and industrial significance, which are key intermediates for multistep synthesis. In addition, we have implemented a number of green strategies in organic transformations using environmental friendly catalytic reaction conditions [34–38]. Herein, we carried out the development of an efficient practical methodology for the



Scheme 1. (Color online.) Synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones.

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